The present invention is in the field of ultraviolet (UV) light source applications and provides transparent acrylic materials for use as optics and covers for such applications. In particular, the present invention provides a UV light engine comprising at least one optical element made of a polymeric material selected from polyalkyl(meth)acrylate and poly(meth)acyralkylimide. The optical element of the ultraviolet light engine may be primary optics, secondary optics, tertiary optics or a combination thereof.
Acrylic materials for use in an ultraviolet light engine

Field of the invention

The present invention is in the field of ultraviolet (UV) light source applications and provides transparent acrylic materials for use as optics and covers for such applications.

In particular, the present invention provides a UV light engine comprising at least one optical element made of a polymeric material selected from the group consisting of polyalkyl(meth)acrylate and poly(meth)acrylalkylimide. The optical element of the UV light engine may be selected from primary optics, secondary optics, tertiary optics or be a combination thereof.

A further aspect of the present invention relates to use of a polymeric material selected from the group consisting of polyalkyl(meth)acrylate and poly(meth)acrylalkylimide as an optical element in a UV light engine. The present invention further provides a process for the manufacturing of a UV light engine, wherein said process comprises at least one step in which polyalkyl(meth)acrylate homo- and copolymers and poly(meth)acrylalkylimide copolymers are used for the manufacturing of an optical element.

In yet a further aspect, the present invention relates to a UV light emitting device, which comprises the UV light engine as described above.

Prior art

UV light sources have been used in several technical areas including e.g. healthcare, lighting (horticultural, agricultural, aquarium), green houses, high colour rendering index (CRI) light sources, counterfeit analysis, curing, disinfection, printing (3D, digital), optical sensors and instrumentation, photocatalytic air purification, lithography etc.

Typical representatives of UV light sources are e.g. mercury vapour lamps, gas-discharge lamps, excimer lasers, plasma or synchrotron sources. More recently, UV light emitting diodes (LEDs) have entered the market, as a result of increasingly available and affordable semiconductors and efficient manufacturing techniques.

Conventional UV light sources often emit light over a broad range of the spectrum. For instance, a typical emission spectrum of a mercury vapour lamp ranges from ~ 180 nm up to ~ 600 nm. One disadvantage of use of such UV light sources is a low effectiveness - only a small part of the emission spectrum is actually required for a specific application.
Furthermore, these UV light sources often have a high operating temperature. Therefore, glass, and in particular quartz glass, are preferred optics materials for UV applications, use of plastic materials is very uncommon.

In combination with mercury vapour lamps optics are usually not used at all because of a typically rather big size of the lamp and high operating temperatures.

In contrast to conventional UV light sources, UV LEDs represent a new artificial UV light source which emit narrow band UV light, and are typically manufactured in a compact way. The effectiveness of UV LEDs in a certain application can thus be much higher, especially when the entire emitted light is optically guided to a certain target area. Since operating temperatures of UV LEDs are significantly lower than in the case of conventional UV light sources, use of cost-efficient optics made of transparent plastic materials becomes possible.

It is well-known that the majority of common transparent polymeric materials such as e.g. polycarbonates have only a limited transmittance of and stability against UV light. The polymer itself, or at least some additives used in the production thereof, can crack and disintegrate when exposed to certain dosages of UV light. This results in a significant yellowing and darkening of the material concomitant with a deterioration of mechanical properties. The lower the wavelength of the UV light, i.e. the higher the energy of the photons, the likelier and the faster is the polymer degradation. Pure polymethylmethacrylate (PMMA) shows a good transmittance in the UVA range (320-400 nm). In practice, however, certain by-products formed during PMMA polymerization, as well as certain processing additives and even some commonly used UV stabilizers, may lead to its degradation or at least yellowing if the material is exposed to UV radiation.

To overcome the problem of limited stability against UV light, commercially available polymeric materials commonly comprise UV absorbers and/or UV absorbing layers. Such materials are described inter alia in patent applications WO 201/0/054905 A1 and WO 201/0/60925 A1. Although these materials show an improved stability against UV light and solar radiation, they cannot be fully utilized in all applications in which a high transmittance in the UV range below 385 nm is required.

CN 103568544 A discloses a curing optical system which comprises an LED array, a fly lens and a convergent lens, wherein a plurality of LEDs are fixed to an LED substrate to form the LED array. CN 103568544 A mentions that the lens material may be glass such as UV or infrared glass or polymethyl methacrylate (PMMA). CN 103568544 A does not teach use of PMMA as a lens material for a UV light device.
US 2016/0076026 A1 describes an UV LED lamp comprising a UV LED chip and a cover, wherein the inner surface and the outer surface of the cover are roughed, and the amount of total reflection from the roughed inner surface is greater than the amount of total reflection from the roughed outer surface. US 2016/0076726 A1 emphasises that if PMMA is used as a cover material, a special PMMA grade having a very low content of additives needs to be employed. The document admits that processability of such PMMA is not as good as processability of conventional polymers and therefore a process for the manufacturing the corresponding cover is more complex than it would be if a conventional PMMA were used.

US 2016/0076726 A1 provides no information in respect to long-term stability of employed PMMA in the claimed UV LED lamp.

**Object of the invention**

Hence, the technical problem addressed by the present invention is to provide a transparent polymeric material for use in an optical element in a UV light engine. Such material needs to have a high transmittance in the target UV wavelength range (here: 350 nm to 400 nm) and an excellent long-term stability against the UV light. Furthermore, such polymeric material should have a sufficient thermal stability at usual operating temperatures of the UV light source.

Furthermore, it is important to ensure that the employed polymeric material can be processed using standard processing techniques for PMMA moulding compounds such as injection moulding or extrusion and no undesired degradation or yellowing of the optical element during the service life time of the UV light engine takes place.

A further technical problem addressed by the present invention was provision of a UV light engine with at least one optical element made of the corresponding transparent polymeric material.

Yet a further technical problem of the present invention was provision of a UV light emitting device comprising the UV light engine as described above.

**Brief description of figures**

Figure 1: schematic illustration of the test rig of the accelerated weathering test:

1. UV LED
2. heated metal block
drilling for test specimen
power supply and driver electronics
water cooled metal baseplate

Summary of the invention

The present invention is based on a surprising finding that compositions comprising polymeric acrylic materials, the transmittance of which at the wave length of 345 nm is not less than 10%, measured at an optical path length of 40 mm, show substantially no signs of undesired yellowing when used in a UV light engine, as long as the employed ultraviolet light source of said UV light engine has an emission peak wavelength between 350 nm and 400 nm. For instance, such compositions show substantially no signs of undesired yellowing after an exposure to monochromatic radiation at 365 nm at an intensity of 288 W/m² for about 4500 h (total radiation dosage of about 5 GJ/m²).

The emission peak half width of the ultraviolet light source is preferably not higher than 30 nm. Ideally, the emission spectrum of the ultraviolet light source comprises a single emission peak.

The corresponding materials (typically PMMA homo- and copolymers or PMMI copolymers in combination with certain stabilizers) have an excellent transmittance in the relevant UV range (350 nm to 400 nm), are substantially haze-free, have a high long-term stability when exposed to narrow band UV radiation and show substantially no signs of yellowing or crack formation. Therefore, these materials can be advantageously used as optical elements in a UV light engine emitting UV light in the range from 350 nm to 400 nm.

Importantly, the employed polymeric materials may comprise common additives such as flow improvers, stabilisers etc. in usual amounts, as long as the transmittance of the polymeric materials at the wave length of 345 nm, measured at an optical path length of 40 mm is not less than 10%. This result is surprising in light of US 2016/0076726 A1 which emphasises that only special PMMA grades having a very low content of additives can be used for this purpose.

Accordingly, the present invention provides a UV light engine comprising at least one UV light source having an emission peak wavelength between 350 nm and 400 nm and, preferably, the emission peak half width not higher than 30 nm and at least one optical element selected from primary optics, secondary optics, tertiary optics or a combination thereof. The optical element may comprise a polymeric material which may be selected from polyalkyl(meth)acrylate and poly(meth)acrylmethylimid.
As will be readily appreciated by a skilled person, the term "polyalkyl(meth)acrylate" as used herein may refer to homo- and copolymers comprising alkyl(meth)acrylate units. The term "poly(meth)acrylmethylimid" typically refers to copolymers.

A further aspect of the present invention relates to the use of polymeric materials selected from the group consisting of polyalkyl(meth)acrylate and poly(meth)acrylmethylimid as an optical element in a UV light engine. In other words, the present invention provides a process for the manufacturing of a UV light engine, the process comprising a step of manufacturing an optical element from a polymeric material selected from the group consisting of polyalkyl(meth)acrylate and poly(meth)acrylmethylimid.

Finally, the present invention provides a UV light emitting device comprising the UV light engine as specified above.

Hence, some aspects of the present invention are as follows:

[1] An ultraviolet light engine comprising:
   at least one ultraviolet light source and
   at least one optical element selected from
   primary optics, secondary optics, tertiary optics or a combination thereof, wherein the optical element comprises a polymeric material selected from the group consisting of polyalkyl(meth)acrylate and poly(meth)acrylmethylimid.

[2] The ultraviolet light engine according to [1], wherein the polyalkyl(meth)acrylate is a copolymer comprising from 80 wt.-% to 99.8 wt.-% of methyl methacrylate units and from 0.2 wt.-% to 20 wt.-% of C_{1-10}-alkyl acrylate units, based on the weight of the copolymer.

[3] The ultraviolet light engine according to [1] or [2], wherein the polyalkyl (meth)acrylate comprises methyl methacrylate units and methyl acrylate units and/or ethyl acrylate units.

[4] The ultraviolet light engine according to [1], wherein the poly(meth)acrylalkylimide has a structure of Formula (I)

\[
\begin{array}{c}
\text{R}^1 \\
\text{O} \\
\text{N} \\
\text{R}^2 \\
\text{O} \\
\text{R}^3 \\
\text{n}
\end{array}
\]

\( (I) \)
in which the moieties R\(^1\) and R\(^2\) are independently a hydrogen atom or a methyl group, R\(^3\) is an alkyl group having from 1 to 20, preferably 1 to 10 carbon atoms.

[5] The ultraviolet light engine according to any of [1] to [4], wherein the polymeric material further comprises from 0 wt.-% to 0.3 wt.-%, preferably from 0 wt.-% to 0.2 wt.-%, more preferably 0.0001 wt.-% to 0.15 wt.-%, more preferably 0.001 wt.-% to 0.1 wt.-%, even more preferably 0.005 wt.-% to 0.05 wt.-% based on the weight of the polymeric material, of a compound of Formula (II):

![Formula (II)](image)

in which the moieties R\(^4\) and R\(^5\) are independently an alkyl or cycloalkyl groups having from 1 to 10 carbon atoms, preferably from 1 to 4 carbon atoms, particularly preferably 2 carbon atoms.

[6] The ultraviolet light engine according to any of [1] to [4], wherein the polymeric material further comprises from 0 wt.-% to 0.3 wt.-%, preferably from 0 wt.-% to 0.2 wt.-%, more preferably 0.0001 wt.-% to 0.15 wt.-%, more preferably 0.001 wt.-% to 0.1 wt.-%, even more preferably 0.005 wt.-% to 0.05 wt.-% based on the weight of the polymeric material, of a compound of Formula (III):

![Formula (III)](image)

in which the moieties R\(^6\) are independently alkyl groups having from 4 to 10 carbon atoms, preferably i-butyl groups, and

the moiety R7 is an alkyl group having from 10 to 20 carbon atoms, preferably n-octadecyl group; and

from 0 wt.-% to 0.3 wt.-%, preferably from 0 wt.-% to 0.2 wt.-%, more preferably 0.0001 wt.-% to 0.2 wt.-%, more preferably 0.001 wt.-% to 0.15 wt.-%, even more preferably 0.005 wt.-% to 0.1 wt.-%, yet even more preferably 0.01 wt.-% to 0.1 wt.-%, based on the weight of the polymeric material, of a compound of Formula (IV):
in which the moiety $R^8$ is an alkyl group having from 1 to 4 carbon atoms, preferably a methyl group.

The ultraviolet light engine according to any of [1] to [4], wherein the polymeric material further comprises from 0 wt.-% to 0.3 wt.-%, preferably from 0 wt.-% to 0.2 wt.-%, more preferably 0.0001 wt.-% to 0.15 wt.-%, more preferably 0.001 wt.-% to 0.1 wt.-%, even more preferably 0.005 wt.-% to 0.05 wt.-% based on the weight of the polymeric material, of a compound of Formula (III):

in which the moieties $R^6$ are independently alkyl group having from 4 to 10 carbon atoms, preferably i-butyl groups and the moiety $R^7$ is an alkyl group having from 10 to 20 carbon atoms, preferably n-octadecyl group; and

from 0 wt.-% to 0.3 wt.-%, preferably from 0 wt.-% to 0.2 wt.-%, more preferably 0.0001 wt.-% to 0.2 wt.-%, more preferably 0.001 wt.-% to 0.15 wt.-%, even more preferably 0.005 wt.-% to 0.1 wt.-%, yet even more preferably 0.01 wt.-% to 0.1 wt.-%, based on the weight of the polymeric material, of a compound of Formula (V):

in which the moiety $R^9$ is an alkyl group having from 1 to 4 carbon atoms, preferably a methyl group and

$R^{10}$ is an alkyne group having from 4 to 16 carbon atoms, preferably an octamethylene group.

The ultraviolet light engine according to any of [1] to [7], wherein the optical element has a yellowness index Y.I., as defined in the norm DIN 6167, measured at an optical path...
length of 40 mm, of not more than 5, preferably not more than 3, measured after accelerated weathering testing under the following conditions:

- optical path length: 40 mm
- sample temperature: 70 °C
- total exposure time: 4 488 h
- irradiance: $288 \pm 17 \text{ W/m}^2$ at 365 nm (peak wavelength; 9 nm spectrum half width) or $555 \pm 41 \text{ W/m}^2$ at 385 nm (peak wavelength; 10 nm spectrum half width) or $429 \pm 31 \text{ W/m}^2$ at 395 nm (peak wavelength; 13 nm spectrum half width).

[9] The ultraviolet light engine according to any of [1] to [8], wherein the optical element has a transmittance of not less than 70%, preferably not less than 80%, more preferably not less than 90% measured in a wavelength interval from 350 nm to 400 nm at an optical path length of 40 mm.

[10] The ultraviolet light engine according to any of [1] to [9], wherein the ultraviolet light source is a light emitting diode (LED), a mercury vapour lamp, an excimer laser, a plasma or synchrotron source or a gas discharge tube.

[11] Use of a polymeric material selected from the group consisting of polyalkyl(meth)acrylate and poly(meth)acrylalkylimide as an optical element in an ultraviolet light engine.

[12] An ultraviolet light emitting device comprising the ultraviolet light engine according to any of [1] to [10].

[13] The ultraviolet light emitting device according to [12], wherein the UV light source is an LED.

**Detailed description of the preferred embodiments**

The term “light engine” as used herein refers to a light module comprising one or several UV light sources and at least one optical element which may form primary optics, secondary optics or tertiary optics of the light engine. Optionally, the UV light engine may also include a thermally conductive element and electronic components other than the UV light sources, e.g. driver electronics. The terms “primary optics”, “secondary optics” and “tertiary optics” are familiar to a skilled person and used in their common meaning. Thus, the term primary
optics refers to a substantially transparent optical component which is attached directly onto a UV light source. Typically, the primary optics are optimized for light extraction, or general beam shaping, or both. Primary optics help the luminous flux of the light source to take a specific radiation pattern. In some embodiments of the present invention, the primary optics are sufficient for distributing the UV light so that the primary optics are the only optical element of the UV light engine.

In a further embodiment, the UV light engine further comprises secondary optics. Secondary optics are attached directly onto or around the UV light source. Typically, the secondary optics are separated from the light source by an air gap, which can also be filled with refractive index matching materials. Secondary optics may be either reflective or refractive, and are used to modify the output beam of a UV light source. Diverging optics spread the incoming light, while collimating optics gather the incoming light into a collimated beam. In some embodiments of the present invention, the secondary optics are sufficient for distributing the UV light so that the secondary optics are the only optical element of the UV light engine.

The UV light engine of the present invention may further comprise tertiary optics. Tertiary optics help to direct, transport or diffuse light emitted by the UV light engine. Common examples of tertiary optics are e.g. light guides, light pipes, diffusers or protective sheets with optical function. By way of example, if tertiary optics are integrated into the design of a UV light engine, there will be no need to re-design the primary and secondary optics if an application requires a different beam angle of UV light. In some embodiments of the present invention, the secondary optics are sufficient for distributing the UV light so that the secondary optics are the only optical element of the UV light engine.

The optical elements of the present invention may comprise a polymeric material selected from the group consisting of polyalkyl(meth)acrylate and poly(meth)acrylalkylimide, wherein the transmittance of the polymeric material at the wave length of 345 nm, measured at an optical path length of 40 mm is not less than 10%. Additionally, it is highly advantageous, that the polymeric material is substantially transparent in the UV light region between 350 nm and 400 nm. For instance, the polymeric material of the optical element may have a transmittance of not less than 40%, preferably not less than 50%, more preferably not less than 60%, even more preferably not less than 70% and particularly preferably not less than 80%, measured at the wavelength of 365 nm at an optical path length of 40 mm.

The transmittance of the polymeric material at the wave length of 345 nm is measured using an instrument as defined in the norm ISO 13468-2 which is suitable for measurements at this wave length (e.g. a Varian Cary 5000). For the measurement a test specimen of the following
dimensions can be used: cross-section: 9.5 x 9.5 mm², length: 40 mm, with the light propagating along the 40 mm axis. The measurement is carried out at 23 °C and 50% RH. Furthermore, the measurement is to be carried out after 96 hours of exposition at the wavelength 365 nm with 288 ± 17 W/m² at the 9.5 x 9.5 mm² cross section at 70 °C. For this purpose, UV LED: Nichia NVSU233A may be used as described in the Examples of the present application. These LEDs are commercially available from the Nichia Corporation (Tokushima, Japan).

According to the present invention, the transmittance of the polymeric material at the wavelength of 345 nm, measured at an optical path length of 40 mm is not less than 10%, more preferably not less than 20%, yet more preferably not less than 30%, particularly preferably not less than 40%. The inventors surprisingly found that the transmittance of the polymeric material at the wave length of 345 nm correlates with the development of the yellowing index during a long-term exposition to UV light having a wavelength between 350 nm and 400 nm. In other words, a sufficiently high transmittance of the polymeric material at the wave length of 345 nm ensures that the material shows substantially no signs of optical degradation or yellowing upon a long-term use in the UV light engine of the present invention. The yellowness index is an accepted indicator for quantifying degradation for clear, colourless plastics. This result is surprising and could not be predicted on the basis of the prior art.

For some other applications, the polymeric material of the optical element may have a transmittance of not less than 40%, preferably not less than 50%, more preferably not less than 60%, even more preferably not less than 70%, and particularly preferably not less than 80%, measured at the wavelength of 385 nm at an optical path length of 40 mm, measured after a 96 h exposure at the wavelength 365 nm 288 ± 17 W/m² at 70 °C.

Depending on the intended purpose, the polymeric material of the optical element may also have a transmittance of not less than 40%, preferably not less than 50%, more preferably not less than 60%, even more preferably not less than 70% and particularly preferably not less than 80%, measured at the wavelength of 395 nm at an optical path length of 40 mm, measured after a 96 h exposure at the wavelength 365 nm 288 ± 17 W/m² at 70 °C.

Preferably, the optical element has a transmittance of not less than 70%, preferably not less than 80%, more preferably not less than 90% in a wavelength interval from 350 nm to 400 nm measured at an optical path length of 40 mm after a 96 h exposure at the wavelength 365 nm 288 ± 17 W/m² at 70 °C. In this context, the expression "transmittance in a wavelength interval from 350 nm to 400 nm" refers to any wavelength in said interval.
All transmittance measurements mentioned in the present application are carried out after 96 hours of exposition at the wavelength 365 nm with 288 ± 17 W/nm at the 9.5 x 9.5 mm² cross section at 70 °C. For this purpose, UV LED: Nichia NVSU233A may be used as described in the Examples of the present application. These LEDs are commercially available from the Nichia Corporation (Tokushima, Japan).

The polyalkyl(meth)acrylate can substantially consist of only one kind of alkyl(meth)acrylate repeating units i.e. be a homopolymer. The polyalkyl(meth)acrylate can moreover be a copolymer of several kinds of alkyl(meth)acrylate monomers. The polyalkyl(meth)acrylate can also be a blend of several different homopolymers or copolymers. The term "(meth)acrylate" as used herein refers not only to methacrylates, e.g. methyl methacrylate, ethyl methacrylate, etc., but also acrylates, e.g. methyl acrylate, ethyl acrylate, etc. and also to mixtures composed of these monomers.

For the purposes of the present invention, particular preference is given to homo- and copolymers of C₁-C₃-alkyl (meth)acrylates, advantageously of C₁-C₄-alkyl (meth)acrylates, in particular of C₁-C₄-alkyl (meth)acrylate polymers, and these can, if appropriate, also comprise monomer units which differ therefrom.

It has proved particularly advantageous to use copolymers which contain from 70% by weight to 100% by weight, in particular from 70% to 100% by weight, of optionally substituted C₁-C₃-alkyl (meth)acrylates. Preferred C₁-C₃-alkyl methacrylates encompass methyl methacrylate, ethyl methacrylate, propyl methacrylate, isopropyl methacrylate, n-butyl methacrylate, isobutyl methacrylate, i-tert-butyl methacrylate, pentyl methacrylate, hexyl methacrylate, heptyl methacrylate, octyl methacrylate, iso-octyl methacrylate, and ethylhexyl methacrylate, nonyl methacrylate, decyl methacrylate, and also cycloalkyl methacrylates, for example cyclohexyl methacrylate, isobornyl methacrylate or ethylcyclohexyl methacrylate. Methyl methacrylate is the particularly preferred C₁-C₃-alkyl (meth)acrylate. C₁-C₃-alkyl may be substituted by one or several hydroxyl groups, wherein unbranched C₁-C₃-0ₕₖₖₘₘ substituents having one primary hydroxyl group are particularly preferred. An example of such (meth)acrylates is 2-hydroxyethyl methacrylate (HEMA).

Preferred C₁-C₃-alkylacrylates encompass methyl acrylate, ethyl acrylate, propyl acrylate, isopropyl acrylate, n-butyl acrylate, isobutyl acrylate, i-tert-butyl acrylate, pentyl acrylate, hexyl acrylate, heptyl acrylate, octyl acrylate, iso-octyl acrylate, nonyl acrylate, decyl acrylate, and ethylhexyl acrylate, and also cycloalkyl acrylates, for example cyclohexyl acrylate, isobornyl acrylate or ethylcyclohexyl acrylate. C₁-C₃-0ₕₖₖₘₘacrylates having one or several
hydroxyl groups may also be employed, wherein acrylates with unbranched \( \text{C}-\text{C}-\text{Q} \text{alkyl} \) substituents having one primary hydroxyl group are particularly preferred. For instance, 2-hydroxyethyl acrylate can be used for this purpose.

5 Very particularly preferred copolymers encompass from 80 wt.-% to 100 wt.-% of methyl methacrylate (MMA) units and from 0 wt.-% to 20 wt.-% of \( \text{C}_1\text{-C}-\text{O}^{\text{alkyl}} \) acrylate units. In yet a more preferred embodiment, the copolymers may encompass from 85 wt.-% to 99.8 wt.-% of methyl methacrylate (MMA) units and from 0.2 wt.-% to 15 wt.-% of \( \text{C}_1\text{-C}-\text{O}^{\text{alkyl}} \) acrylate units. In a most preferred embodiment, the copolymers may encompass from 90 wt.-% to 99.8 wt.-% of methyl methacrylate (MMA) units and from 0.2 wt.-% to 10 wt.-% of \( \text{C}_1\text{-C}-\text{O}^{\text{alkyl}} \) acrylate units.

C\(\text{-C}-\text{O}^{\text{alkyl}} \) acrylate units are preferably selected from methyl acrylate units, ethyl acrylate units and/or butyl acrylate units. In a particularly preferred embodiment, the polyalkyl(meth)acrylate comprises methyl methacrylate units and methyl acrylate units and/or ethyl acrylate units. The corresponding copolymers are commercially available from Evonik Performance Materials GmbH under the PLEXIGLAS\textsuperscript{®} trademark on the European, Asian, African and Australian continents and under the ACRYLITE\textsuperscript{®} trademark in North America and South America.

The polyalkyl(meth)acrylates can be produced by polymerization processes, and particular preference is given here to free-radical polymerization processes, in particular bulk polymerization, solution polymerization, suspension polymerization and emulsion polymerization processes. Initiators particularly suitable for these purposes encompass in particular azo compounds, such as 2,2'-azobis(isobutynitrile) or 2,2'-azobis(2,4-dimethylvaleronitrile), redox systems, e.g. the combination of tertiary amines with peroxides or sodium disulphite and persulphates of potassium, sodium or ammonium, or preferably peroxides (in which connection cf. for example H. Rauch-Puntigam, Th. Volker, "Acryl- und Methacrylverbindungen" [Acrylic and methacrylic compounds], Springer, Heidelberg, 1967, or Kirk-Othmer, Encyclopedia of Chemical Technology, Vol. 1, pages 386 ff, J. Wiley, New York, 1978). Examples of particularly suitable peroxide polymerization initiators are di lauroyl peroxide, ieri-butyl peroctoate, ieri-butyl perisonanoate, dicyclohexyl peroxodicarbonate, dibenzoyl peroxide and 2,2-bis(tert-buty perox)butane. It is also possible and preferred to carry out the polymerization reaction using a mixture of various polymerization initiators of different half-lifetime, examples being dilauroyl peroxide and 2,2-bis(ieri-butylperoxy)butane, in order to maintain a constant stream of free radicals during the course of the polymerization reaction, and also at various polymerization temperatures. The
amounts used of polymerization initiator are generally from 0.01 wt.-% to 2 wt.-%, based on the weight of the monomer mixture.

The polymerization reaction can be carried out continuously or batch wise. After the polymerization reaction, the polymer is obtained by way of conventional steps of isolation and separation. For instance, when PMMA is produced by suspension polymerization and emulsion polymerization processes filtration, coagulation and spray drying can be employed.

The chain lengths of the polymers or copolymers can be adjusted by polymerizing the monomer or monomer mixture in the presence of molecular-weight regulators, a particular example being the mercaptans known for this purpose, e.g. n-butyl mercaptan, n-dodecyl mercaptan, 2-mercaptoethanol or 2-ethylhexyl thioglycolate, pentaerythritol tetrathioglycolate; the amounts used of the molecular-weight regulators generally being from 0.05 wt.-% to 5 wt.-%, preferably from 0.1 wt.-% to 2 wt.-% and particularly preferably from 0.2 wt.-% to 1 wt.-%, based on the weight of the monomer or monomer mixture (cf. H. Rauch-Puntigam, Th. Volker, “Acryl- und Methacrylverbindungen” [Acrylic and methacrylic compounds], Springer, Heidelberg, 1967; Houben-Weyl, Methoden der organischen Chemie [Methods of organic chemistry], Vol. XIV/1, page 66, Georg Thieme, Heidelberg, 1961, or Kirk-Othmer, Encyclopedia of Chemical Technology, Vol. 1, pages 296ff, J. Wiley, New York, 1978). n-Dodecyl mercaptan is particularly preferably used as a molecular-weight regulator.

In a further embodiment the optical element may comprise poly(meth)acrylalkylimide copolymers. The poly(meth)acrylalkylimide copolymers comprise structural unit which may be represented by the following Formula (I):

![Formula (I)](image)

in which the moieties $R^1$ and $R^2$ are independently a hydrogen atom or a methyl group, $R^3$ is an alkyl group having from 1 to 20, preferably 1 to 10 carbon atoms. In a particularly preferred embodiment, the moieties $R^1$, $R^2$ and $R^3$ are methyl groups, $n$ is typically from 60 to 6000, more preferably from 100 to 2000.

The monomeric units in the structure (I) preferably form more than 30 wt.-%, particularly preferably more than 50 wt.-% and very particularly preferably more than 80 wt.-% of the
poly(meth)acrylalkylimide copolymer, based on the total weight of the monomers. The molecular weight Mw of the poly(meth)acrylalkylimide copolymers typically range from about 50 000 g/mol to about 250 000 g/mol, in particular from about 80 000 g/mol to about 120 000 g/mol.

In addition, poly(meth)acrylalkylimide copolymers may contain further monomeric units which arise, for example, from esters of acrylic or methacrylic acid, in particular with lower alcohols having 1-4 carbon atoms, styrene, maleic acid or the anhydride thereof, itaconic acid or the anhydride thereof, vinylpyrrolidone, vinyl chloride or vinylidene chloride. The proportion of the comonomers, which cannot be cyclized or can be cyclized only with very great difficulty, should not exceed 30 wt.-%, preferably 20 wt.-% and particularly preferably 10 wt.-%, based on the weight of the monomers.

The preparation of poly(meth)acrylalkylimide copolymers is known and is disclosed, for example, in GB 1 078 425, GB 1 045 229, DE 1 817 156 or DE 27 26 259.

The materials of the optical element are preferably those which comprise poly(V-methylmethacrylimides) (PMMI) and/or polymethyl methacrylates (PMMA). Poly(V-methylmethacrylimides) (PMMI), polymethyl methacrylates (PMMA) and/or PMMI-PMMA copolymers are preferably copolymers of PMMI and PMMA which are prepared by partial cycloimidization of PMMA. PMMI which is prepared by partial imidization of PMMA is usually prepared in such a way that not more than 83 wt.-% of the PMMA used are imidized. The resulting product is referred to as PMMI but strictly speaking is a PMMI-PMMA copolymer. Both PMMA and PMMI or PMMI-PMMA copolymers are commercially available, for example from Evonik Performance Materials GmbH under the PLEXIMID® trademark on the European, Asian, African and Australian continents and under the ACRYMID® trademark in North America and South America. The products and their preparation are known (Hans R. Kricheldorf, Handbook of Polymer Synthesis, Part A, published by Marcel Dekker Inc. New York - Basel - Hong Kong, page 223 et seq.; H.G. Elias, Makromolekule [Macromolecules], published by Huthig und Wepf Basel - Heidelberg - New York; US 2 146 209 and US 4 246 374).

In some embodiments of the present invention, provided the transmittance of the polymeric material at the wave length of 345 nm, measured at an optical path length of 40 mm is not less than 10%, the material of the optical element further comprises at least one compound of general Formula (II)
in which the moieties $R^4$ and $R^5$ are independently an alkyl or cycloalkyl groups having from 1 to 10 carbon atoms, preferably from 1 to 4 carbon atoms, particularly preferably 2 carbon atoms.

Among the preferred alkyl groups are the methyl, ethyl, propyl, isopropyl, 1-butyl, 2-butyl, 2-methylpropyl, ieri-buty1, pentyl, 2-methylbutyl, 1,1-dimethylpropyl, hexyl, heptyl, octyl, 1,1,3,3-tetramethylbutyl, nonyl, 1-decyl and 2-decyl group. Among the preferred cycloalkyl groups are the cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl and cyclooctyl group, which optionally have branched or unbranched alkyl groups as substituents.

Preference is given to use of the compound of the Formula (IIa)

![Formula IIa](image)

This compound is available commercially from the BASF SE as Tinuvin® 312.

Typically, the compounds of Formula (II) are employed as UV absorbers and therefore the materials comprising the compounds of Formula (II) have a low transmittance in the UV region. The inventors surprisingly found that when the material of the optical element comprises not more than 0.3 wt.-% of a compound of general Formula (II), preferably not more than 0.2 wt.-%, more preferably not more than 0.15 wt.-%, more preferably not more than 0.1 wt.-%, even more preferably not more than 0.05 wt.-%, even more preferably not more than 0.01 wt.-%, even more preferably not more than 0.001 wt.-%, based on the weight of the material of the optical element, the resulting material still has a good transmittance in a selected, relevant UV region and an excellent long-term stability in the UV light. Therefore, such material can be advantageously used in an optical element of a UV light engine.

The polymeric material preferably comprises not less than 0.0001 wt.-% of a compound of general Formula (II), more preferably not less than 0.001 wt.-%, based on the weight of the polymeric material. This ensures that the polymeric material can be exposed to UV light for longer periods of time without any signs of yellowing or degradation of mechanical
properties. In this context, it is worth mentioning that, depending on other constituents, the polymeric material such as PMMA may sometimes still have an acceptable stability upon exposure to UV light if it does not comprise the compound of the Formula (II) at all.

In view of the above, it can be readily appreciated that the polymeric material for use in the present invention may comprise from 0 wt.-% to 0.3 wt.-% of a compound of general Formula (II), more preferably from 0 wt.-% to 0.2 wt.-%, more preferably 0.0001 wt.-% to 0.15 wt.-%, more preferably 0.001 wt.-% to 0.1 wt.-%, even more preferably 0.005 wt.-% to 0.05 wt.-%, based on the weight of the polymeric material as long as the transmittance of the polymeric material at the wavelength of 345 nm, measured at an optical path length of 40 mm is not less than 10%.

In a further embodiment of the present invention, the polymeric material of the optical element comprises a compound of Formula (II):

\[
\text{HO} \quad \text{R}^6 \quad \text{OR}^7 \quad \text{R}^8
\]

(III)

in which the moieties \( R^6 \) are independently alkyl groups in combination with a compound of Formula (IV):

\[
\text{OH} \quad \text{OR}^8
\]

(IV)

in which the moiety \( R^8 \) is an alkyl group having from 1 to 4 carbon atoms.

Although the compounds of Formula (III) and Formula (IV) have a relatively low absorption in the region between 350 nm and 400 nm, the polymeric material comprising a combination of these two compounds has an excellent stability against UV radiation and shows substantially no signs of yellowing or darkening upon a long-term exposure to UV light. For this reason, polyalkyl(meth)acrylate or poly(meth)acrylalkylimide comprising the combination of compounds of Formulae (II) and (IV) are highly suitable as an optical element in a UV light engine.

For the sake of achieving an optimal optical performance of the material, it is preferable that it comprises not more than 0.3 wt.-%, more preferably not more than 0.2 wt.-%, more preferably not more than 0.15 wt.-%, yet even more preferably not more than 0.1 wt.-%, and particularly preferably not more than 0.05 wt.-% of the compound of Formula (II). The
content of the compound of Formula (IV) is typically chosen to be not higher than 0.3 wt.-%, preferably not higher than 0.2 wt.-%, preferably not higher than 0.15 wt.-%, particularly preferably not higher than 0.1 wt.-% and even more preferably not higher than 0.05 wt.-%, based on the total weight of the polymeric material.

On the other hand, a particularly high long-time resistance of the polymeric material against UV radiation is achieved when it comprises more than 0 wt.-%, preferably not less than 0.0001 wt.-%, even more preferably not less than 0.001 wt.-% and particularly preferably not less than 0.005 wt.-% of the compound of Formula (III). The content of the compound of Formula (IV) is commonly chosen to be more than 0 wt.-%, preferably not less than 0.0001 wt.-%, yet even more preferably not less than 0.001 wt.-%, even more preferably not less than 0.005 wt.-% and yet even more preferably not less than 0.01 wt.-%.

Thus, in the corresponding embodiment the polymeric material preferably comprises from 0 wt.-% to 0.3 wt.-%, preferably from 0 wt.-% to 0.2 wt.-%, more preferably 0.0001 wt.-% to 0.15 wt.-%, more preferably 0.001 wt.-% to 0.1 wt.-%, even more preferably 0.005 wt.-% to 0.05 wt.-% based on the weight of the polymeric material, of a compound of Formula (II), and from 0 wt.-% to 0.3 wt.-%, preferably from 0 wt.-% to 0.2 wt.-%, more preferably 0.0001 wt.-% to 0.2 wt.-%, more preferably 0.001 wt.-% to 0.15 wt.-%, even more preferably 0.005 wt.-% to 0.1 wt.-%, yet even more preferably 0.01 wt.-% to 0.1 wt.-%, based on the weight of the polymeric material, of a compound of Formula (IV).

In the compound of Formula (II):

![Chemical structure](III)

the moieties \( R^6 \) may be identical or different from each other and are independently alkyl groups having from 4 to 10 carbon atoms. The moieties \( R^6 \) may be represented by 1-butyl, 2-butyl, 2-methylpropyl, ieri-butyl, pentyl, 2-methylbutyl, 1,1-dimethylpropyl, hexyl, heptyl, octyl, 1,1,3,3-tetramethylbutyl, nonyl, 1-decyl, or 2-decyl groups, wherein ieri-butyl groups are preferred.

The moiety \( R^7 \) may be an alkyl group having from 10 to 20 carbon atoms. Examples thereof are e.g. 1-decyl, 2-decyl, undecyl, dodecyl, pentadecyl, octadecyl and eicosyl group, the \( n \)-octadecyl group being particularly preferred.
Preference is given to use of the compound of Formula (Iia):

![Chemical structure](image)

This compound is commercially available from BASF SE under the product name IRGANOX® 1076.

In the compound of Formula (IV):

![Chemical structure](image)

the moiety $R^8$ is an alkyl group having from 1 to 4 carbon atoms. Among the preferred alkyl groups are the methyl, ethyl, propyl, isopropyl, 1-butyl, 2-butyl, 2-methylpropyl and 1-ethylbutyl.

In a particularly preferred embodiment, $R^8$ is a methyl group and the compound of Formula (IV) is methyl salicylate.

In yet a further embodiment of the present invention the polymeric material comprises a compound of Formula (III) as described above in combination with a compound of Formula (V):

![Chemical structure](image)

in which the moieties $R^9$ are independently from each other alkyl groups having from 1 to 4 carbon atoms and $R^{10}$ is an alkylene group having from 4 to 16 carbon atoms. The moieties $R^9$ may be identical or different from each other. The moieties $R^9$ may be represented by methyl, ethyl, propyl, isopropyl, 1-butyl, 2-butyl, 2-methylpropyl or 1-ethylbutyl, methyl groups being particularly preferred. $R^{10}$ may be butylene, 2-methylpropylene, pentylene, 2-methylbutylene, 1,1-dimethylpropylene, hexamethylene, heptamethylene, octamethylene, 1,1,3,3-tetramethylbutylene, nonamethylene, decamethylene, undecamethylene, dodecamethylene, pentadecamethylene or hexadecamethylene. Preferably, $R^{10}$ is an octamethylene group.
Preference is given to use of the compound of Formula (Va):

This compound is commercially available from BASF SE under the product name TINUVIN® 770.

Again, although the compounds of Formula (II) and Formula (V) have a relatively low absorption in the region between 350 nm and 400 nm, the polymeric material comprising a combination of these two compounds has an excellent stability against UV radiation between 350 nm and 400 nm and shows substantially no signs of optical degradation such as yellowing or crack formation upon a long-term exposure to UV light. For this reason, polyalkyl(meth)acrylate or poly(meth)acrylalkylimide comprising a combination of the compounds of Formulae (II) and (V) is highly suitable as an optical element in the UV light engine of the present invention.

For the sake of achieving an optimal optical performance of the material, it is preferable that it comprises not more than 0.1 wt.-%, more preferably not more than 0.08 wt.-%, yet even more preferably not more than 0.05 wt.-% of the compound of Formula (III), based on the total weight of the polymeric material. The content of the compound of Formula (V) is advantageously chosen to be not higher than 0.3 wt.-%, preferably not higher than 0.2 wt.-%, particularly preferably not higher than 0.15 wt.-% and even more preferably not higher than 0.1 wt.-%, based on the total weight of the polymeric material.

Applicants believe that a particularly high long-time resistance of the polymeric material against UV radiation in the wave lengths between 350 nm and 400 nm is achieved when it comprises more than 0 wt.-%, preferably not less than 0.0001 wt.-%, even more preferably not less than 0.001 wt.-% and particularly preferably not less than 0.005 wt.-% of the compound of Formula (III). The content of the compound of Formula (V) is typically chosen to be more than 0 wt.-%, preferably not less than 0.0001 wt.-%, yet even more preferably not less than 0.01 wt.-%, even more preferably not less than 0.05 wt.-% and yet even more preferably not less than 0.1 wt.-%.
Thus, in the corresponding embodiment the polymeric material preferably comprises from 0 wt.-% to 0.3 wt.-%, preferably from 0 wt.-% to 0.2 wt.-%, preferably 0.0001 wt.-% to 0.15 wt.-%, more preferably 0.001 wt.-% to 0.1 wt.-%, even more preferably 0.005 wt.-% to 0.05 wt.-% based on the weight of the polymeric material, of a compound of Formula (II), and from 0 wt.-% to 0.3 wt.-%, preferably from 0 wt.-% to 0.2 wt.-%, preferably 0.0001 wt.-% to 0.2 wt.-%, more preferably 0.001 wt.-% to 0.15 wt.-%, even more preferably 0.005 wt.-% to 0.1 wt.-%, yet even more preferably 0.01 wt.-% to 0.1 wt.-% of a compound of Formula (V), based on the weight of the polymeric material.

The compounds of Formulae (II)-(V) can be incorporated in the polymeric material of the optical element by methods that are known from the literature, for example by mixing with the polymer prior to further processing at higher temperature, by addition to the polymer melt or by addition to the suspended or dissolved polymer while it is being processed. They can optionally also be added to the starting materials for production of the polymeric material, and they do not lose their stabilizing effect even in the presence of other usual light and heat stabilizers, oxidizing and reducing agents and the like. Hence, the compounds of Formulae (II)-(V) become uniformly distributed in the matrix of polyalkyl(meth)acrylate or poly(meth)acrylalkylimide.

A polymeric material that is especially preferred for the purposes of the present invention has a softening temperature of not less than 80°C (Vicat softening temperature VST (ISO 306-B50)). It is therefore particularly suitable as an optical element in a UV light engine, as it does not begin to creep as a result of increased operating temperatures during use.

As further possibility, the monomeric alkyl(meth)acrylates and the compounds of Formulae (II)-(V) may be admixed to a polymerizable monomeric mixture (casting monomer mixture). Casting monomer mixtures comprise, within the scope of the present invention, both mixtures of monomers and mixtures of monomers, polymers and oligomers, so-called syrup or resin mixtures. Optical elements can be produced from the casting monomer mixtures by known methods, preferably cell cast polymerization and continuous casting polymerization.

In addition to the previously mentioned additives, within the scope of the present invention it may optionally be useful to employ further additives that are well known by a person skilled in the art, as long as these additives do not significantly lower the UV transmittance of the polymeric material in the desired region. External lubricants, antioxidants, flame retardants, further hindered amine light stabilizers (HALS), flow improvers such as stearyl alcohol or palmitic acid, metal additives for screening against electromagnetic radiation, antistatic
agents, mould-release agents, dyes, pigments, adhesion promoters, anti-weathering agents, plasticizers, fillers and the like are preferred.

Within the scope of an especially preferred embodiment of the present invention, at least one sterically hindered amine may be used, giving a further improvement in resistance to weathering. Yellowing or degradation of materials exposed to external conditions for a long time can be further reduced.

Especially preferred sterically hindered amines include dimethylsuccinate-1 -(2-hydroxyethyl)-4-hydroxy-2,2,6,6-tetramethylpiperazine polycondensate, poly[(6-(1,1,3,3-tetramethylbutyl)amino-1 ,3,5-triazine-2,4-diyl][(2,2,6,6-tetramethyl-4-piperidyl)iminoo]-hexamethylene((2,2,6,6-tetramethyl-4-piperidyl)imino)], A/A'-bis(3-aminopropyl)ethylene-diamine-2,4-bis[V-butyl-A/-{1,2,2,6,6-pentamethyl-4-piperidyl]amino]-6-chloro-1 ,3,5-triazine condensate, bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate and 2-(3,5-di-t-4-hydroxybenzyl)-2-n-butylmalonate bis(1 ,2,2,6,6-pentamethyl-4-piperidyl).

Furthermore, the use of silane adhesion promoters or organic titanium compounds has proved quite especially useful, giving further improvement in adhesion to inorganic materials. Suitable silane adhesion promoters include vinyltrichlorosilane, vinyl-tris(P-methoxyethoxy)silane, vinyltriethoxysilane, vinyltrimethoxysilane, γ-methacryloxy-propyltrimethoxysilane, 3-(3,4-epoxycyclohexyl)ethyltrimethoxysilane, γ-glycidoxypropyl-methyldiethoxysilane, A/-3-(aminoethyl)- γ-aminopropyltrimethoxysilane, A/-3-(aminoethyl)- γ-aminopropylmethyldimethoxysilane, γ-amino-propyltriethoxysilane, A/-phenyl-γ-amino-propyltrimethoxysilane, γ-mercaptopropyltrimethoxysilane and γ-chloropropyltrimethoxysilane.

Especially advantageous optical elements are those produced from polymeric materials and/or casting monomer mixtures that possess a sufficiently high UV transmittance. Over the wavelength range from 350 nm to 400 nm the transmittance is preferably at least 40%, more preferably at least 50%, more preferably at least 60%, even more preferably at least 70%, yet even more preferably not less than 80%, most preferably not less than 90% (measurement using the Cary 5000 spectrophotometer from the company Agilent (formerly Varian) at an optical path length of 40 mm). In this context, the transmittance is defined as the arithmetic mean of spectral transmittance in the wavelength range from 350 nm to 400 nm, measured after a 96 h exposure at the wavelength 365 nm 288 ± 17 W/m² at 70 °C.
As a result of its excellent UV stability against UV light having a wavelength between 350 nm and 400 nm, the optical element for use in the present invention has a yellowness index Y.I., as defined in the norm DIN 6167 (D65/1 0°) and determined with a Varian Cary 5000 spectrophotometer, of not more than 5, preferably not more than 4, more preferably not more than 3, measured after accelerated weathering test for 4488 h under the conditions described in the section "Examples".

The choice of the UV light source for use in the UV light engine of the present invention is not particularly limited, as long as the ultraviolet light source has an emission peak wavelength between 350 nm and 400 nm. The emission peak half width of the UV light source is typically not higher than 30 nm, preferably not higher than 20 nm, more preferably not higher than 15 nm and the operating temperature of the light source is compatible with the employed polymeric material. Ideally, the UV light source is substantially monochromatic i.e. has a single emission peak. The examples of suitable light sources include a light emitting diode (LED), an excimer laser, a plasma or synchrotron source or a gas discharge tube. As already discussed above, use of LEDs is particularly preferred.

In a further aspect, the present invention relates to use of a polymeric material selected from the group consisting of polyalkyl(meth)acrylate and poly(meth)acrylalkylimide as an optical element in a UV light engine having an emission peak wavelength between 350 nm and 400 nm.

In still a further aspect the invention provides a UV light emitting device comprising the UV light engine as described above and having an emission peak wavelength between 350 nm and 400 nm. Ideally, the emission spectrum of the ultraviolet light source comprises a single emission peak and its emission peak half width is not higher than 30 nm, more preferably not higher than 20 nm, even more preferably not higher than 15 nm. Said UV light emitting device can be manufactured at lower costs and has a considerably higher mechanical stability in comparison to a UV light emitting device which employs inorganic glass or quartz glass in its optical elements.

The following examples illustrate the invention in a greater detail. However, there is no intention that the present invention be restricted to these examples.

Examples

Tested Polymeric Materials

The tested polymeric material (PMMA) was prepared by mixing of 98.92 wt.-% of methyl methacrylate, 1.00 wt.-% of methyl acrylate, 0.04 wt.-% of dilauroyl peroxide and 0.04 wt.-
% of n-dodecyl mercaptan, and heating to 60 °C for 36 hours. The polymerisate was crushed by aid of a polymer mill and further processed on an extruder with degassing unit and feeding the following additives:

Example 1 (comparative): The tested PMMA with 100 ppm of the compound of Formula (Ma) and 800 ppm of stearyl alcohol.

Example 2: The tested PMMA with 550 ppm of the compound of Formula (Ilia) and 880 ppm of methyl salicylate and 1000 ppm of stearyl alcohol.

Example 3: The tested PMMA with 1000 ppm of stearyl alcohol.

Example 4: The tested PMMA with 1200 ppm of the compound of Formula (Ilia) and 1000 ppm of compound of Formula (Va) and 1000 ppm of stearyl alcohol.

Example 5 (comparative): The tested PMMA with 800 ppm of the compound of Formula (Ma) and 500 ppm of palmitic acid.

The resulting mixtures were pelletized and testing specimens in the dimensions 9.5 x 9.5 x 40 mm³ were manufactured by aid of press moulding, followed by cutting and polishing. The materials of Examples 1-5 are also suitable for processing using other standard processing techniques for PMMA moulding compounds such as injection moulding or extrusion.

Stability Testing

The specimens were placed in a UV LED light engine composed of a water cooled metal block with UV LEDs attached, a heated metal block with openings for the specimens, power supply, electronics and drivers, and a lid (Figure 1). The UV LEDs test rigs were operated in a laboratory kept at 23 °C and 50% RH. The tests were carried out in the air atmosphere at atmospheric pressure and an air circulation was provided during the entire test.

The UV light enters the test specimen of the dimensions: cross-section: 9.5 x 9.5 mm², length: 40 mm, light propagating along the 40 mm axis. A Qmini radiometer from the company RGB Photonics was used to determine the UV irradiance received by the acrylic specimens (wide configuration, integral between 350 nm and 450 nm) and to further follow the degradation rate of the UV LEDs. An instrument as defined in the norm ISO 13468-2 (a Varian Cary 5000 spectrophotometer) was used to measure the spectral transmittance and yellowness index
(Y.I.) along the 40 mm axis after certain time intervals.

Accelerated weathering testing was performed under the following conditions:

- Optical path length: 40 mm
- Sample temperature: 70°C
- Total exposure time: 4488 hours
- 365 nm UV LED: Nichia NVSU233A
  UV irradiance received by specimen: \( \sim 288 \pm 17 \text{ W/nm}^2 \) (350 nm - 450 nm; arithmetic mean average irradiance of 4 UV LEDs and over the entire testing time)
- 385 nm UV LED: Nichia NVSU233A
  UV irradiance received by specimen: \( \sim 555 \pm 41 \text{ W/nm}^2 \) (350 nm - 450 nm; arithmetic mean average irradiance of 4 UV LEDs and over the entire testing time)
- 395 nm UV LED: Nichia NSVU233A
  UV irradiance received by specimen: \( \sim 429 \pm 31 \text{ W/nm}^2 \) (350 nm - 450 nm; arithmetic mean average irradiance of 4 UV LEDs and over the entire testing time)

**Results**

**Optical transmittance**

Optical measurements of the specimens reveal excellent transmittance properties of the polymeric materials at tested UV LED wavelengths.

To evaluate suitability of the tested polymeric materials for use as optical elements at wavelengths of 345 nm, 365 nm, 385 nm and 395 nm, the corresponding transmittance values were determined.

Table 1 summarizes the transmittance values of the tested samples at 345 nm after exposition to UV light having an emission maximum at 365 nm for 96 h:

<table>
<thead>
<tr>
<th>Example</th>
<th>Transmittance, % at 345 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.7</td>
</tr>
<tr>
<td>2</td>
<td>45.0</td>
</tr>
<tr>
<td>3</td>
<td>83.3</td>
</tr>
<tr>
<td>4</td>
<td>63.2</td>
</tr>
<tr>
<td>5</td>
<td>0.0</td>
</tr>
</tbody>
</table>

Table 1. Transmittance at 365 nm of tested polymeric materials after exposition to UV light having an emission maximum at 365 nm for 96 h.

Table 2 summarizes the transmittance values of the tested samples at 365 nm:
Table 2. Transmittance at 365 nm of tested polymeric materials after exposition to UV light having an emission maximum at 365 nm.

Table 3 summarizes the transmittance values of the tested samples at 385 nm:

<table>
<thead>
<tr>
<th>Irradiation time, h</th>
<th>Transmittance, % at 365 nm</th>
<th>Transmittance, % at 385 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Example 1</td>
<td>Example 2</td>
</tr>
<tr>
<td>0</td>
<td>52.9</td>
<td>78.7</td>
</tr>
<tr>
<td>24</td>
<td>57.7</td>
<td>88.1</td>
</tr>
<tr>
<td>48</td>
<td>57.9</td>
<td>88.1</td>
</tr>
<tr>
<td>96</td>
<td>58.5</td>
<td>89.2</td>
</tr>
<tr>
<td>192</td>
<td>58.3</td>
<td>89.1</td>
</tr>
<tr>
<td>288</td>
<td>58.3</td>
<td>89.2</td>
</tr>
<tr>
<td>384</td>
<td>57.7</td>
<td>88.6</td>
</tr>
<tr>
<td>504</td>
<td>57.9</td>
<td>88.7</td>
</tr>
<tr>
<td>840</td>
<td>57.4</td>
<td>88.1</td>
</tr>
<tr>
<td>1176</td>
<td>57.4</td>
<td>87.7</td>
</tr>
<tr>
<td>1512</td>
<td>56.9</td>
<td>87.3</td>
</tr>
<tr>
<td>2016</td>
<td>56.9</td>
<td>87.0</td>
</tr>
<tr>
<td>2520</td>
<td>56.8</td>
<td>87.1</td>
</tr>
<tr>
<td>3192</td>
<td>56.4</td>
<td>87.1</td>
</tr>
<tr>
<td>3864</td>
<td>56.4</td>
<td>87.3</td>
</tr>
<tr>
<td>4488</td>
<td>56.6</td>
<td>87.2</td>
</tr>
</tbody>
</table>
Table 3. Transmittance at 385 nm of tested polymeric materials after exposition to UV light having an emission maximum at 385 nm.

Table 4 summarizes the transmittance values of the tested samples at 395 nm:

<table>
<thead>
<tr>
<th>Irradiation time, h</th>
<th>Transmittance, % at 395 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Example 1</td>
</tr>
<tr>
<td>0</td>
<td>89.0</td>
</tr>
<tr>
<td>24</td>
<td>91.3</td>
</tr>
<tr>
<td>48</td>
<td>90.9</td>
</tr>
<tr>
<td>96</td>
<td>91.4</td>
</tr>
<tr>
<td>192</td>
<td>91.6</td>
</tr>
<tr>
<td>288</td>
<td>91.5</td>
</tr>
<tr>
<td>384</td>
<td>91.5</td>
</tr>
<tr>
<td>504</td>
<td>91.7</td>
</tr>
<tr>
<td>840</td>
<td>91.9</td>
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<tr>
<td>1176</td>
<td>91.9</td>
</tr>
<tr>
<td>1512</td>
<td>91.7</td>
</tr>
<tr>
<td>2016</td>
<td>91.7</td>
</tr>
<tr>
<td>2520</td>
<td>91.8</td>
</tr>
<tr>
<td>3192</td>
<td>91.6</td>
</tr>
<tr>
<td>3864</td>
<td>91.5</td>
</tr>
<tr>
<td>4488</td>
<td>91.5</td>
</tr>
</tbody>
</table>

Table 4. Transmittance at 395 nm of tested polymeric materials after exposition to UV light having an emission maximum at 395 nm.

The sample with a high concentration of the compound of Formula (Ma) (Example 5) does not provide for sufficient transmittance for a 365 nm UV LED, however, it performs reasonably well with the e.g. 395 nm UV LED, and is additionally most stable towards outdoor weathering. All other specimens exhibit transmittance values of 85% and above for the target wavelengths of the UV LEDs.

The test results summarized in Tables 2-4 indicate that the polymeric materials of Examples 1-4 show high transmittance values at wavelength of 365 nm. Furthermore, the materials...
show no significant decrease of transmittance at this wavelength. Consequently, the materials can be advantageously used in an optical element of a UV light engine operating at this wavelength.

Irradiation with UV light at a wavelength of 365 nm shows a stronger effect on the transmittance of the tested materials than irradiation with UV light having longer wavelengths. This effect is observed in a visual comparison of the specimens, changes are expressed as the change of yellowness index (Y.I., D65/1° according to DIN 6167) before and after testing (Y.I.448 h – Y.I.0 h) summarized in Table 5.

Table 5 summarizes the changes in yellowness indices of the tested samples at 365 nm, 385 nm and 395 nm:

<table>
<thead>
<tr>
<th>Irradiation with</th>
<th>Δ Y.I. (4 488 hours – 0 hours) (Y.I. at 0 hours)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Example 1</td>
</tr>
<tr>
<td>365 nm LED</td>
<td>1.63 (0.81)</td>
</tr>
<tr>
<td>385 nm LED</td>
<td>-0.41 (0.81)</td>
</tr>
<tr>
<td>395 nm LED</td>
<td>-0.43 (0.81)</td>
</tr>
</tbody>
</table>

Table 5. Yellowness Index (D65/1°) of tested polymeric materials after exposition to UV light.

All polymeric materials of Examples 1-5 show a drop in yellowness index during the first 96 hours of irradiation and become more transparent in the visible region. After 4488 hours of testing all samples except for Example 1 and Example 5, both equipped with the compound of Formula (Ia), still have a yellowness index below the starting value. Example 4, equipped with the compounds of Formula (IIa) and Formula (Va) shows an especially remarkable drop in yellowness index with a continuous downward tendency over the entire testing time.

From Table 1 can be noted that the polymeric materials of Examples 1 and 5 have a transmittance at the wavelength of 345 nm below 10%. As a result, both materials develop a significant yellowing index after a long-term exposure to UV radiation having a wavelength of 365 nm. These results are surprising, because materials of Examples 1 and 5 comprise the UV-absorber Tinuvin 312 (the compound of Formula (Ia)) and therefore could be expected to have a higher UV stability and show a lower yellowing index after a long-term exposure to UV radiation.

The composition of Example 4 shows substantially no signs of yellowing after a long-term exposure to UV radiation. However, the overall performance of the compositions in
Examples 2 and 3 is slightly better than in Example 4, because the composition of Example 4 has a slightly lower transmission at the wavelength of 365 mm.

Importantly, all tested samples showed no signs of mechanical degradation.

It is surprising that the optical properties of the tested PMMA samples of Examples 2-4 remain at quite high constant transmittance values for the given wavelengths, even after exposure of several thousands of hours at a testing temperature of 70°C. In contrast thereto, the compositions of Examples 1 and 5 shows some yellowing after a long-term exposure to UV light having a wavelength of 365 mm.
Claims

1. An ultraviolet light engine comprising:
   at least one ultraviolet light source and
   at least one optical element selected from
   primary optics, secondary optics, tertiary optics or a combination thereof, wherein the
   optical element comprises a polymeric material selected from the group consisting of
   polyalkyl(meth)acrylate and poly(meth)acrylalkylimide and wherein
   the transmittance of the polymeric material at the wavelength of 345 nm, measured at
   an optical path length of 40 mm is not less than 10% and
   the ultraviolet light source has an emission peak wavelength between 350 nm and 400
   nm.

2. The ultraviolet light engine according to Claim 1, wherein the polyalkyl(meth)acrylate is
   a copolymer comprising from 80 wt.-% to 100 wt.-% of methyl methacrylate units and
   from 0 wt.-% to 20 wt.-% of C₁-C₄ alkyl acrylate units, based on the weight of the
   copolymer.

3. The ultraviolet light engine according to Claim 1 or 2, wherein the polyalkyl
   (meth)acrylate comprises methyl methacrylate units and methyl acrylate units and/or
   ethyl acrylate units.

4. The ultraviolet light engine according to Claim 1, wherein the poly(meth)acrylalkylimide
   has a structure of Formula (I)

   \[ \begin{array}{c}
   \text{R}^1 \\
   \text{R}^2 \\
   \text{R}^3 \\
   \end{array} \]

   in which the moieties \( \text{R}^1 \) and \( \text{R}^2 \) are independently a hydrogen atom or a methyl group,
   \( \text{R}^3 \) is an alkyl group having from 1 to 20, preferably 1 to 10 carbon atoms.

5. The ultraviolet light engine according to any of Claims 1 to 4, wherein the polymeric
   material further comprises from 0 wt.-% to 0.3 wt.-%, preferably from 0 wt.-% to 0.2 wt.-%
   , more preferably 0.0001 wt.-% to 0.15 wt.-%, more preferably 0.001 wt.-% to 0.1 wt.-%
   , even more preferably 0.005 wt.-% to 0.05 wt.-% based on the weight of the
   polymeric material, of a compound of Formula (II):
in which the moieties $R^4$ and $R^5$ are independently an alkyl or cycloalkyl groups having from 1 to 10 carbon atoms, preferably from 1 to 4 carbon atoms, particularly preferably 2 carbon atoms.

5 6. The ultraviolet light engine according to any of Claims 1 to 4, wherein the polymeric material further comprises
from 0 wt.-% to 0.3 wt.-%, more preferably from 0 wt.-% to 0.2 wt.-%, preferably 0.0001 wt.-% to 0.15 wt.-%, more preferably 0.001 wt.-% to 0.1 wt.-%, even more preferably 0.005 wt.-% to 0.05 wt.-% based on the weight of the polymeric material, of a compound of Formula (II):

$$
\begin{align*}
\text{(II)}
\end{align*}
$$

in which the moieties $R^6$ are independently alkyl groups having from 4 to 10 carbon atoms, preferably i-butyl groups, and
the moiety $R^7$ is an alkyl group having from 10 to 20 carbon atoms, preferably $n$-octadecyl group; and

from 0 wt.-% to 0.3 wt.-%, more preferably from 0 wt.-% to 0.2 wt.-%, more preferably 0.0001 wt.-% to 0.3 wt.-%, more preferably 0.001 wt.-% to 0.15 wt.-%, even more preferably 0.005 wt.-% to 0.1 wt.-%, yet even more preferably 0.01 wt.-% to 0.1 wt.-%, based on the weight of the polymeric material, of a compound of Formula (IV):

$$
\begin{align*}
\text{(IV)}
\end{align*}
$$

in which the moiety $R^8$ is an alkyl group having from 1 to 4 carbon atoms, preferably a methyl group.

7. The ultraviolet light engine according to any of Claims 1 to 4, wherein the polymeric material further comprises
from 0 wt.-% to 0.3 wt.-%, more preferably from 0 wt.-% to 0.2 wt.-%, preferably 0.0001 wt.-% to 0.15 wt.-%, more preferably 0.001 wt.-% to 0.1 wt.-%, even more preferably 0.005 wt.-% to 0.05 wt.-% based on the weight of the polymeric material, of a compound of Formula (II):

$$
\begin{align*}
\text{(II)}
\end{align*}
$$

in which the moieties $R^4$ and $R^5$ are independently an alkyl or cycloalkyl groups having from 1 to 10 carbon atoms, preferably from 1 to 4 carbon atoms, particularly preferably 2 carbon atoms.
0.005 wt.-% to 0.05 wt.-% based on the weight of the polymeric material, of a compound of Formula (III):

\[
\text{HO-} \quad \text{R}^6 \quad \text{R}^6 \quad \text{O}^7 \quad \text{OR}^7 \quad (\text{III})
\]

in which the moieties R^6 are independently alkyl group having from 4 to 10 carbon atoms, preferably i-butyl groups and the moiety R^7 is an alkyl group having from 10 to 20 carbon atoms, preferably n-octadecyl group; and

from 0 wt.-% to 0.3 wt.-%, more preferably from 0 wt.-% to 0.2 wt.-%, preferably 0.0001 wt.-% to 0.2 wt.-%, more preferably 0.001 wt.-% to 0.15 wt.-%, even more preferably 0.005 wt.-% to 0.1 wt.-%, yet even more preferably 0.01 wt.-% to 0.1 wt.-%, based on the weight of the polymeric material, of a compound of Formula (V):

\[
\text{\text{H-N}} \quad \text{R}^9 \quad \text{R}^9 \quad \text{O-R}^{10} \quad \text{O-R}^9 \quad \text{NH} \quad \text{R}^9 \quad \text{R}^9 \quad (\text{V})
\]

in which the moiety R^9 is an alkyl group having from 1 to 4 carbon atoms, preferably a methyl group and R^10 is an alkyene group having from 4 to 16 carbon atoms, preferably an octamethylene group.

8. The ultraviolet light engine according to any of Claims 1 to 7, wherein the optical element has a yellowness index Y.I., as defined in the norm DIN 6167, measured at an optical path length of 40 mm, of not more than 5, preferably not more than 3, measured after accelerated weathering testing under the following conditions:

- optical path length: 40 mm
- sample temperature: 70 °C
- total exposure time: 4488 h
- irradiance: 288 ± 17 W/m² at 365 nm (peak wavelength; 9 nm spectrum half width) or 555 ± 41 W/m² at 385 nm (peak wavelength; 10 nm spectrum half width) or 429 ± 31 W/m² at 395 nm (peak wavelength; 13 nm spectrum half width).
9. The ultraviolet light engine according to any of Claims 1 to 8, wherein the optical element has a transmittance of not less than 70%, preferably not less than 80%, more preferably not less than 90% in a wavelength interval from 350 nm to 400 nm measured at an optical path length of 40 mm.

10. The ultraviolet light engine according to any of Claims 1 to 9, wherein the ultraviolet light source is a light emitting diode (LED), an excimer laser, a plasma or synchrotron source or a gas discharge tube.

11. Use of a polymeric material selected from the group consisting of polyalkyl(meth)acrylate and poly(meth)acrylalkylimide as an optical element in an ultraviolet light engine.

12. An ultraviolet light emitting device comprising the ultraviolet light engine according to any of Claims 1 to 10.

13. The ultraviolet light emitting device according to Claim 12, wherein the UV light source is an LED having an emission peak half width not higher than 30 nm, preferably not higher than 20 nm, more preferably not higher than 15 nm.
## INTERNATIONAL SEARCH REPORT

### A. CLASSIFICATION OF SUBJECT MATTER

INV. C08K5/134 C08K5/20 C08K5/3435 G02B1/04

ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

### B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C08K G02B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data

### C. DOCUMENTS CONSIDERED TO BE RELEVANT

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Date of the actual completion of the international search: 11 October 2017

Date of mailing of the international search report: 19/10/2017

Name and mailing address of the ISA:
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016

Authorized officer: Masson, Patrick

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